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RADIOLOGICAL HEALTH

HASL-235

PLUTONIUM IN SOIL AROUND THE ROCKY FLATS PLANT

August 1, 1970

Health and Safety Laboratory USAEC
New York, New York

UNITED STATES ATOMIC ENERGY COMMISSION • DIVISION OF TECHNICAL INFORMATION

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HASL-235
HEALTH AND SAFETY (TID-4500)

PLUTONIUM IN SOIL AROUND THE ROCKY FLATS PLANT

by

P. W. Krey and E. P. Hardy

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U. S. Atomic Energy Commission
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SUMMARY OF FINDINGS

Soil samples were collected to a depth of 20 cm at 33 sites extending as far as 40 miles from the Dow Chemical Co.'s Rocky Flats plant in Colorado. Deposition concentrations of Pu-239 as high as 2000 mCi/km^2 were found off the plant site but these high concentrations decreased rapidly with distance. The contamination pattern extends eastward from the plant in the direction of the resultant wind vector and has virtually no westward component. The pattern is incompatible with the wind direction on the day of the May 11, 1969 fire. Leaking barrels of plutonium laden cutting oil stored in the southeast corner of the plant are considered the likely source of the contaminant.

Three mCi/km^2 of Pu-239 is the lowest contour readily discernible in the contamination pattern and extends about 8 miles east and southeast of the plant. The inventory of Rocky Flats Pu-239 within the 3 mCi/km^2 contour but excluding AEC controlled land is 2.6 Ci. The extent of the Rocky Flats Pu-239 beyond this contour is difficult to determine because the deposition of Pu-239 from nuclear weapon tests fallout is not precisely known in the Denver area. Our best estimate of the cumulative fallout Pu-239 is 1.5 mCi/km^2 based upon a 1965 soil sample from nearby Derby, Colorado. The most distant sites at 40 miles to the east and north of the plant show a slightly higher value of 2.0 mCi/km^2. The intervening sites average 2.4 mCi/km^2. If the extreme and unlikely assumption is made that the entire area lying 40 miles to the east and north of the plant is contaminated with 1 mCi/km^2, an additional 3.2 Ci of Rocky Flats plutonium could have been released to these remote areas. Sr-90 analyses of the Rocky Flats soils and other studies are in process which might resolve this uncertainty in the remote areas, but additional sampling may be required to thoroughly solve the problem.

The plutonium has been shown to move down into the soil as far as 13 cm, although the distribution shows wide variability apparently depending upon soil chemistry. In several cases there is as much as 60% of the total plutonium below 5 cm. The analytical precision of aliquoting the sample and analysis was shown to be ±20%. Analyses of duplicate samples taken from the same location showed similar precision indicating that the soil sampling was representative of the area studied.

INTRODUCTION

On May 11, 1969 a serious fire broke out at the Atomic Energy Commission's plutonium processing plant at Rocky Flats, Colorado[1]. Stimulated by the possible release of plutonium to the environment by this fire, Dr. Martell demonstrated the presence of Rocky Flats plutonium in soil around the plant [2]. On further investigation it appeared that there were three other plausible sources for this off-site plutonium, i.e.:

1. the September 11, 1957 fire;
2. leaking drums of contaminated cutting oil in a storage area in the southeast corner of the plant complex;
3. a chronic low level stack effluent.

Because of our expertise in soil sampling for radionuclides and their analyses, HASL was invited to perform an initial study of the distribution of Pu-239 in soil around the plant. The objectives of this initial study were to:

(a) confirm and broadly quantify the levels of Rocky Flats plutonium in the soil;

(b) define the maximum distances from the plant at which significant Rocky Flats plutonium could be detected; and

(c) if possible, determine the quantity of Rocky Flats plutonium in the soil off-site, that is, off AEC property.

Preoperational Survey

To assist us in designing a sampling program, we requested Dr. I. Van der Hoven, Chief, ERL, ESSA, Washington, D.C. to make a meteorological examination of the Rocky Flats area. In response C. R. Dickson and G. E. Start surveyed the area during the week of January 26, 1970, and in their memorandum to the files [3] made the following observations and recommendations:

1. More if not most of the Rocky Flats plutonium in the soil around the plant was contributed by the 1957 fire and the leakage from the contaminated drums.

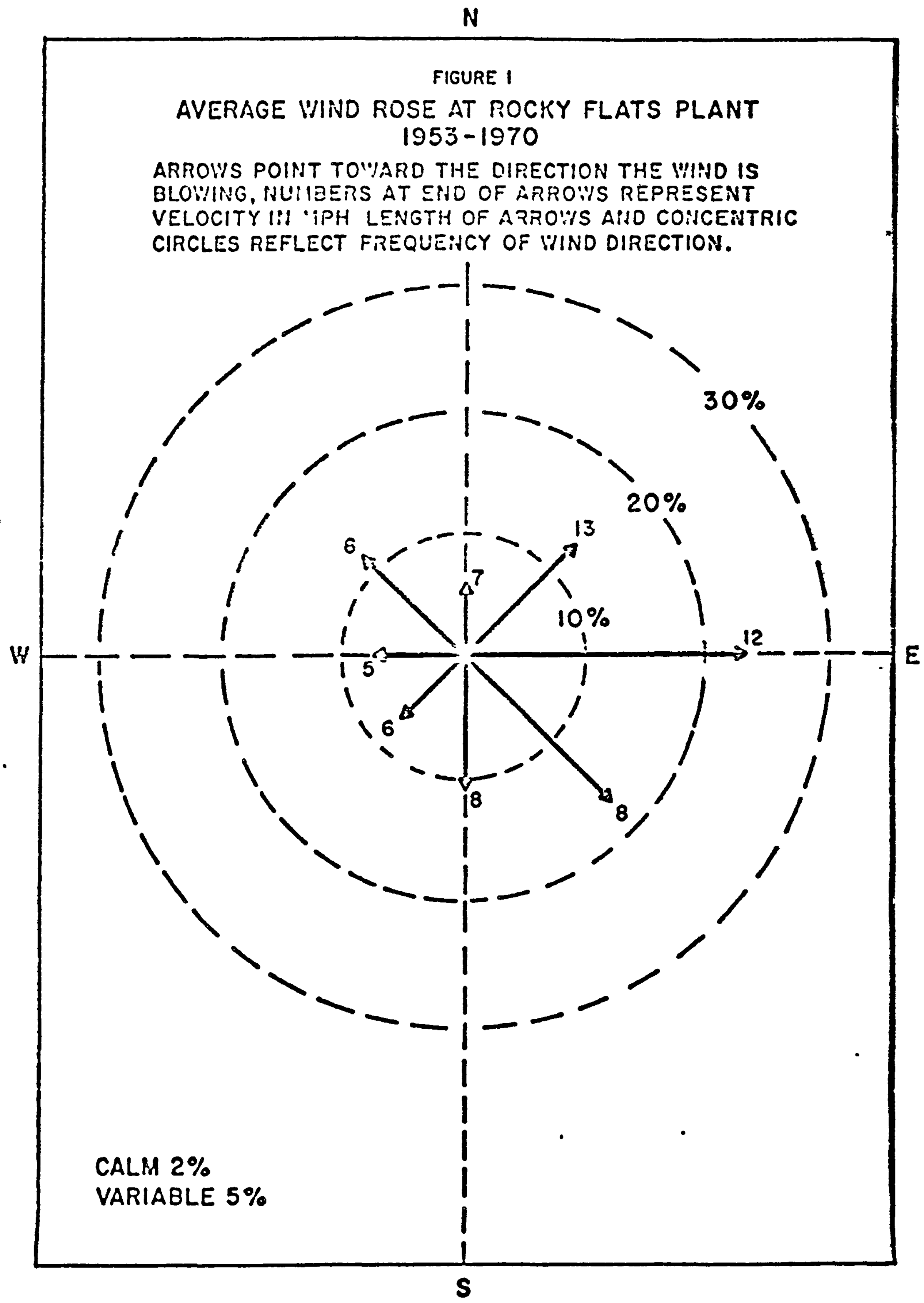
2. These drums were placed in outside storage from 1958 until 1968. Initial leakage was detected in 1964. The change in the distribution of this contamination on the plant property during a period of one year suggested that a gradual southeastward movement may have taken place.

3. The wind rose averaged over a 17-year interval at the Rocky Flats plant is given in Figure 1. West winds occur almost 25% of the time; over 50% of the winds have a westerly component. The strongest gusts and the directions of the most frequent strong gusts (speeds greater than 40 mph) are from the west. The resultant wind vector (derived from the wind rose frequencies and mean speeds) points almost exactly eastward.

FIGURE 1

AVERAGE WIND ROSE AT ROCKY FLATS PLANT
1953-1970

ARROWS POINT TOWARD THE DIRECTION THE WIND IS BLOWING, NUMBERS AT END OF ARROWS REPRESENT VELOCITY IN MPH LENGTH OF ARROWS AND CONCENTRIC CIRCLES REFLECT FREQUENCY OF WIND DIRECTION.



The Denver wind rose shows that southerly to southwesterly winds are most frequent. Air flowing eastward from Rocky Flats should merge with the southerly air flow across Denver and be carried north-north eastward. The South Platte River Valley, about 20 miles east of Rocky Flats, is the logical terrain feature along which this transition from westerly flow should approach completion.

The wind direction on 5/11/69, the day of the last serious fire, was mostly from the north - northeast at speeds from 2 - 10 mph. Figure 2 gives mean hourly winds during that fire. The wind directions during the 9/11/57 fire are shown in Fig. 3[4].

4. Strong gusty winds frequently occur in the lee of the mountains at Rocky Flats; several days each year wind gusts exceed 70 to 80 mph. Consequently, wind erosion has removed much of the fine soil and alluvium leaving mostly rock and sand particles at the ground surface. Two creeks, Walnut and Woman Creeks, pass to the north and south of the plant in an easterly direction. These creeks begin to flow through significantly deep gullies (several hundred yards wide) as they flow by the eastern limits of the plant. The soil in the gullies which was wind eroded from the flats is finer and deeper, and is more likely to become a trap for windborne particles of plutonium.

FIGURE 2

AVERAGE HOURLY WINDS DURING FIRE OF MAY 11, 1969

ARROWS POINT TOWARD THE DIRECTION THE AVERAGE HOURLY WINDS WERE BLOWING. THE LENGTH OF THE ARROWS AND THE CONCENTRIC CIRCLES REFLECT THE WIND VELOCITY IN MPH.

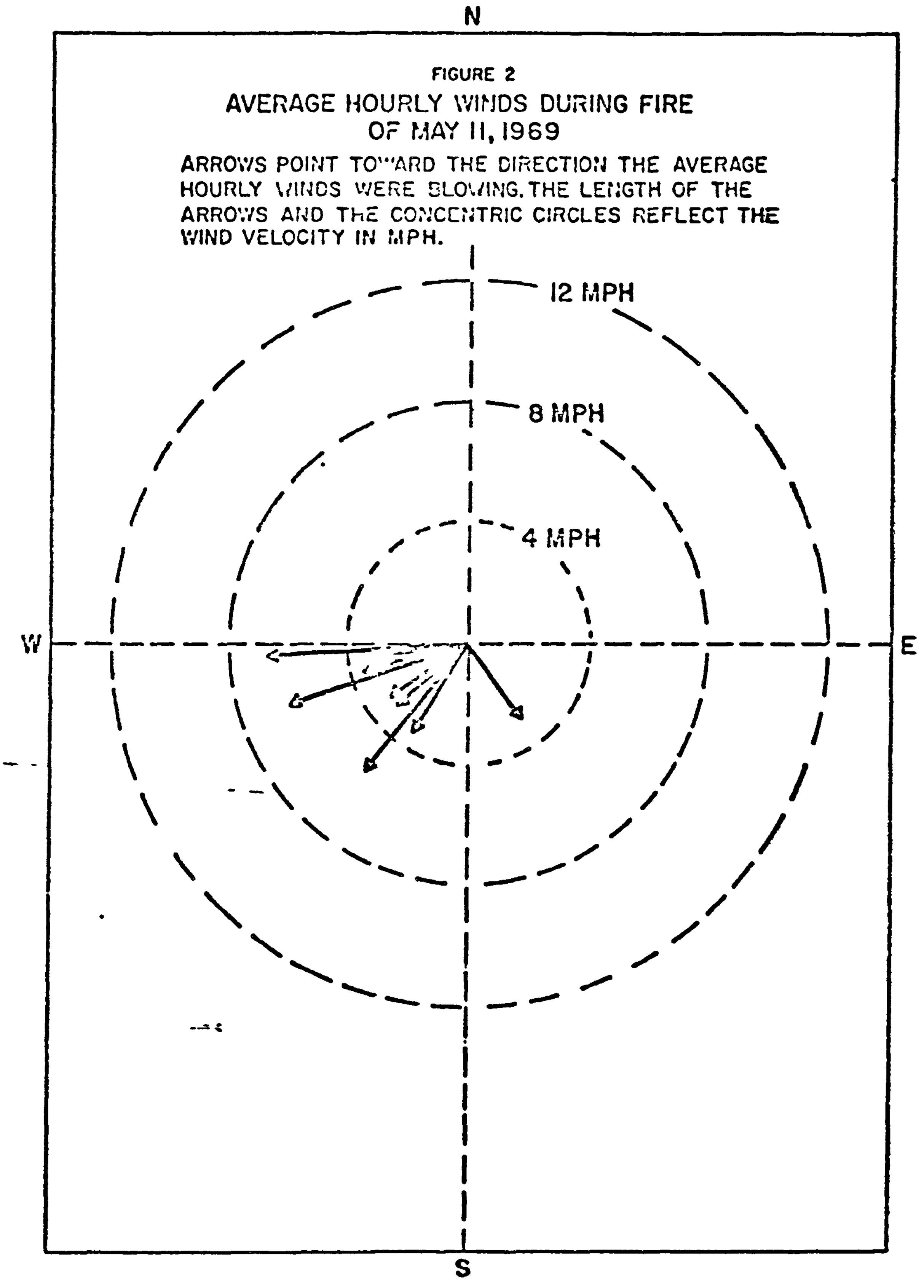
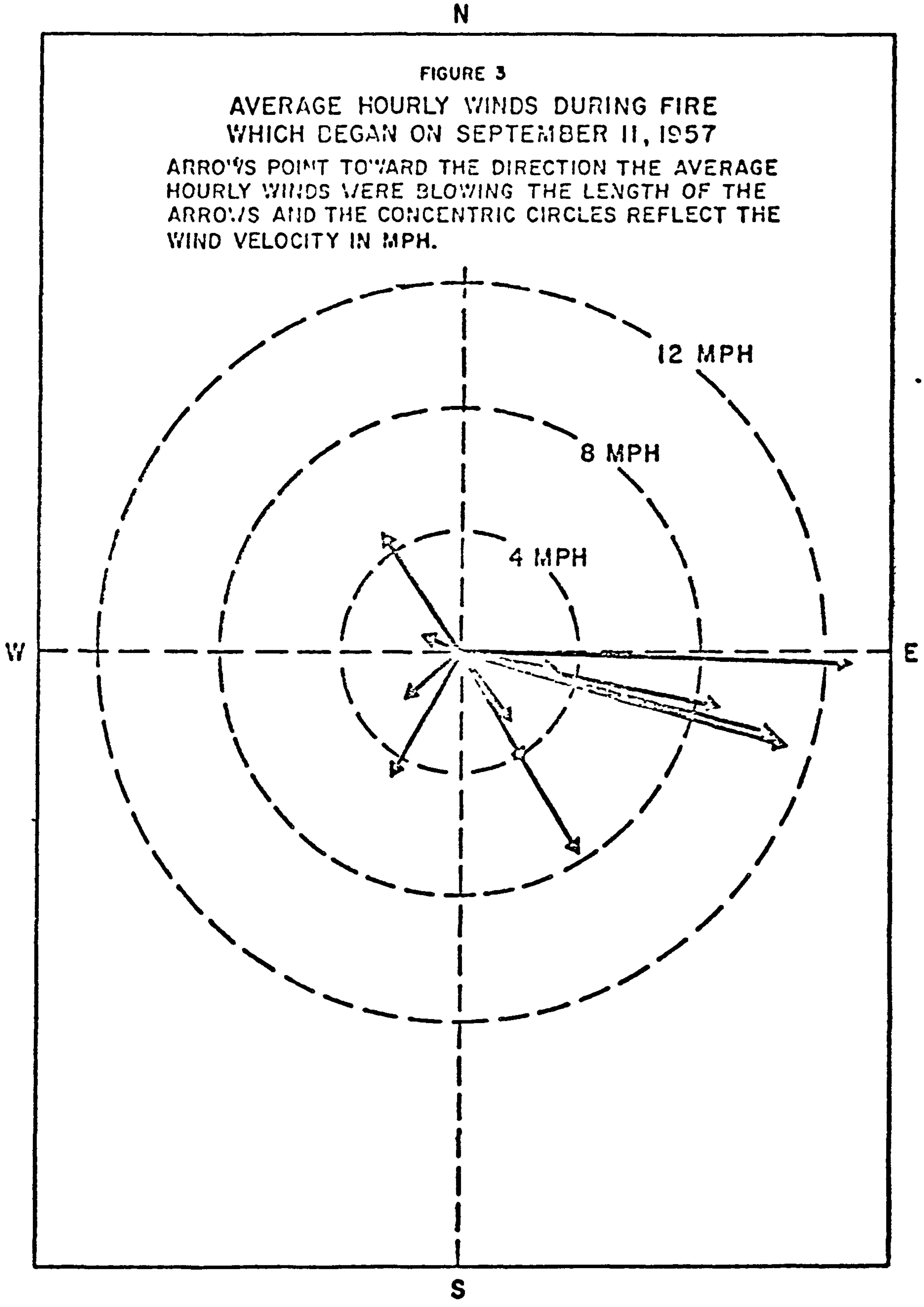


FIGURE 3

AVERAGE HOURLY WINDS DURING FIRE WHICH BEGAN ON SEPTEMBER 11, 1957

ARROWS POINT TOWARD THE DIRECTION THE AVERAGE HOURLY WINDS WERE BLOWING THE LENGTH OF THE ARROWS AND THE CONCENTRIC CIRCLES REFLECT THE WIND VELOCITY IN MPH.



Sampling Site Location

The week of February 9, 1970 was set aside for the collection of the soil samples. Based upon the pre-operational survey, emphasis was given to site location in the downwind direction from the plant. If the results of this limited one week sampling revealed serious deficiencies, the intent was to collect additional samples later to satisfy the need. The selected sampling sites and their description are given in Table 1 and plotted in Figure 4.

SOIL SAMPLING

The sampling techniques used in the Rocky Flats area were primarily dependent upon the rockiness of the soil. In those areas where rocks did not pose a problem, we used the core sampling method[5]. In the immediate vicinity of the Rocky Flats plant, core sampling was not possible. Here it was necessary to cut out a one square foot area sample to the required depth. We tried to select sites for sampling that had been undisturbed since the plant started operations in 1953. The sites were vegetated and in large flat open areas where it could reasonably be assumed that deposited material would not have been removed by water erosion. We avoided sites where debris might have accumulated such as in depressions, at the foot of slopes, or in flooded areas. Our objective was to take samples that would

HASL SOIL SAMPLING SITES

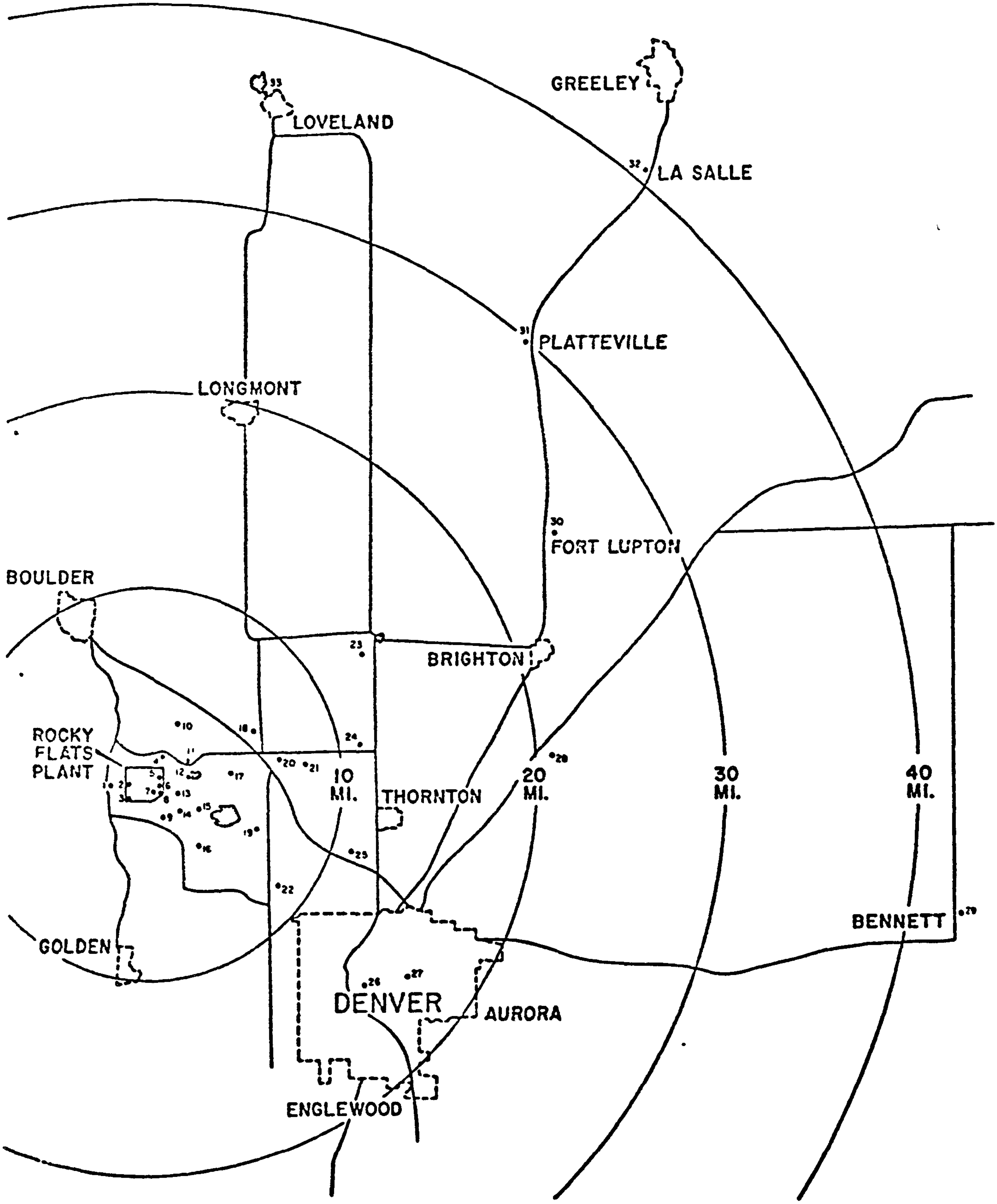
Table 1

near DOW CHEMICAL CO., ROCKY FLATS PLANT, COLORADO

Map Site	Sample	Area and Depth of Sample	Site Description
1	1	12" x 12½" x 8" deep	Church property No 9
2	2	12½" x 13" x 8" deep	Dow property - near west fence
3	3	12½" x 12½" x 8" deep	Dow property - near west fence
4	4	10, 3½" d cores, 0-8"	Off Rt 128
5	5	10, 3½" d cores, 0-6"	Dow property - high ridge near east fence
6	6	12" x 12" x 8" deep	Dow property - near east fence
	7	12½" x 12½" x 8" deep	duplicate of sample 6
	8	12" x 12" x 2" deep	Dow property - near east fence
	9	2 - 3"	sub-soil of above
	10	3 - 4"	" " " "
	11	4 - 5"	" " " "
	12	5 - 6"	" " " "
	13	6 - 7"	" " " "
7	14	10, 3½" d cores, 0-8"	Dow property - near Woman Creek
	15	10 3½" d cores, 0-8"	duplicate of sample 14
8	16	10, 3½" d cores, 0-2"	Dow property - Woman Creek near east fence
	17	10, 3½" d cores 2-8"	sub-soil of above
	18	1 4" d core, 0-2½"	Dow property - Woman Creek near east fence
	19	1 4" d core 2½" - 3½"	sub-soil of above
	20	1, 3" d core, 3½" - 5"	" " " "
	21	1, 3½" d core, 5" - 8"	" " " "
9	22	12" x 12" x 8" deep	Church property No. 23
10	23	12" x 12½" x 8" deep	Hardaway property No 25
11	24	10, 3½" d cores, 0-8"	off Rt 128
12	25	10, 3½" d cores, 0-2"	Great Western Reservoir
	26	10, 3½" d cores, 2-8"	sub-soil of above
	27	1 3½" d core, 0 - ~4"	Great Western Reservoir
13	28	12½" x 12½" x 8" deep	Church property No 13
14	29	10, 3½" d cores, 0-2"	Church property No. 13
	30	10 3½" d cores, 2-8"	sub soil above
15	31	12" x 12" x 8" deep	S E corner of Zehnder property No 18
16	32	10, 3½" d cores, 0-8"	Alkire and 82nd Ave
17	33	12½" x 12½" x 8" deep	108 Ave and Simms
18	34	10 3½" d cores, 0-8"	Swann Hereford Ranch
19	35	10, 3½" d cores, 0-8"	Calkins Lake near W 88th Ave.
20	36	12½" x 12½" x 8" deep	[redacted] E of Jefferson A P near Wadsworth
21	37	10, 3½" d cores, 0-8"	Sheridan and 112th Ave. Murray property
22	38	10, 3½" d cores, 0-8"	Seacrest El.Sch, Quay @ 64th Ave
23	39	10, 3½" d cores, 0-8"	Roy Baxter Farm, Huron and 160th Ave.
24	40	10, 3½" d. cores, 0-8"	Wesley Chapel Cemetery, Huron & 120th Ave.
25	41	10, 3½" d cores, 0-8"	Rausch El Sch. Pecos and Elmwood
26	42	10 3½" d cores, 0-2"	Denver Civic Center
	43	10, 3½" d cores, 2-8"	sub-soil of above
27	44	10, 3½" d cores, 0-2"	Denver City Park
	45	10, 3½" d cores, 2-8"	sub-soil of above
28	46	10, 3½" d cores, 0-8"	Kellson Farm, 120th Ave and Chambers
29	47	10, 3½" d cores, 0-4"	Walt Schroth Farm, Bennett, Colo.
	48	10, 3½" d cores, 4-8"	sub-soil of above
30	49	11, 3½" d cores, 0-8"	Town Park, Ft Lupton, Colo.
31	50	10, 3½" d cores, 0-8"	Town Park, Platteville, Colo.
32	51	10, 3½" d cores, 0-8"	Sylvester Farm, LaSalle, Colo.
33	52	10, 3½" d cores, 0-8"	Lakeside Park, Loveland, Colo.

Figure 4

HASL SOIL SAMPLING SITES NEAR DOW CHEMICAL CO., ROCKY FLATS PLANT



measure the total amount of plutonium that had fallen out. At the time, we arbitrarily decided to sample from the surface down to 20 centimeters, expecting that at this depth we would include all the plutonium. We shall briefly describe the sampling techniques used.

In those areas where core sampling was possible, we first laid out a straight line transect about 5 meters long. With about 30 cm (one foot) spacings in between cores, we cut out ten 8.9 cm (3.5 in.) diameter plugs from the surface to 5 cm, including the vegetation and root mat. In this way we sampled an area of 622 cm^2 (0.67 ft^2). Next we removed the soil from 5 to 20 cm with an auger which bores an 8.9 cm diameter hole. At some sites, we separated the 0 - 5 cm and 5 - 20 cm samples. In most cases, however, the two fractions were combined. Further sampling details and descriptions of the tools are given elsewhere[5].

At map site No. 8, we sampled a one hole depth profile by mechanically drilling with hole saws to a depth of 13 cm. Sampling from 13 to 20 cm was done with the hand auger.

At those sites where there was a preponderance of rocks in the soil, the preferred method of core sampling could not be done. Instead we placed a 930 square centimeter (one square foot) metal template on top of the spot selected for sampling and cut out the sample with chisels and scoops. The rocks were included with the sample.

At map site No. 6, we took samples of soil at various depths down to 20 cm. This was a very rocky area so that we had to use the one square foot template and chisel out the samples as carefully as possible. The first 5 cm were relatively easy to sample but as we went down in depth it was necessary to taper toward the center of the 930 cm^2 area to avoid contaminating with soil from the upper levels. Consequently we could not get an accurate area measurement below 5 cm. To convert from the measured concentrations (dpm per gram) to activity per unit area we assumed a soil density of 1.2 grams per cm^3 down to 15 cm and 2.4 grams per cm^3 from 15 to 18 cm. These density values were obtained as averages of a considerable body of data accumulated at HASL over a 15 year period. As we sampled the depth profile at this site, we removed and discarded the rocks.

Ten of the Rocky Flats samples required the removal of the larger rocks. We assumed that an insignificant amount of plutonium would be in the rock fraction and consequently based the calculated activity per unit area on the weight of the soil fraction. To be sure this was proper, we analyzed the separated rock fraction of the soil containing the highest concentration of plutonium. We found only 0.2 percent of the total plutonium associated with the separated rocks and therefore neglected the rock fraction in the remaining samples.

RADIOCHEMICAL ANALYSIS

Sample Preparation

When the soil samples arrived at HASL, they were spread out on plastic sheets and allowed to air dry for several days. During the drying process, we broke up soil aggregates and pulled apart the top soil plugs consisting of vegetation and root mat. We cut up the vegetation so that it could be distributed homogeneously throughout the sample. When the sample was completely dry it was weighed in its entirety. Then, when necessary, any rocks were removed and weighed separately.

The sample was then crushed and transferred to a blender. After mixing, we spread the sample out on a piece of plastic, marked off quarters, and took scoop fulls from each quarter in a consecutive manner until about a kilogram had been collected. This sub-sample was then pulverized in a rotary hammer mill and transferred to a polyethylene bottle.

One hundred gram aliquots of the pulverized material were aliquoted at various times for submission to the analytical laboratory. This soil sample preparation procedure and equipment used for crushing, blending, and pulverizing are described elsewhere [5].

Radiochemical Procedure

The radiochemical procedure for plutonium in soil was developed by Norton Chu at the Health and Safety Laboratory. It involves leaching a 100 gram aliquot of soil with hydrochloric and nitric acids. We show below that this method effectively removes all the plutonium. Plutonium is isolated by anion exchange (nitrate form) and electrodeposited on a platinum disc. The plutonium isotopes are resolved by alpha spectrometry and yielded with Pu-236. This procedure is described in detail elsewhere [5].

Most of the analyses of the Rocky Flats soils were done by two contractor laboratories: Teledyne Isotopes of Palo Alto, California (designated here as IPA) and Trapelo Division West of Richmond, California (TLW).

Complete dissolution of a sample for radiochemical analysis is the classical and most dependable method of analysis. However, in the case of soil, it is also most difficult, tedious, and time consuming. The HASL leach method was developed to eliminate the need for complete dissolution but to provide accurate, dependable results. To show that the leach method fulfilled its objectives, many samples were analyzed by both leaching and complete dissolution and gave comparable results.

Three different samples from Rocky Flats were selected as representing widely different soil types. Duplicate aliquots of each were analyzed for Pu-239 by the leach and fusion methods. The fusion method affords complete dissolution of the sample by decomposing it in molten sodium carbonate. Table 2 gives the results of these analyses and shows that the mean percent deviation between the methods was 13% with no obvious bias.

Other soils were similarly analyzed and the results are also reported in Table 2. A second method of complete dissolution was used for some of these samples, and is indicated as HF dissolution in the table. This technique affects complete dissolution by prolonged digestion of the sample with hudrofluoric and perchloric acids. The mean percent deviation between methods for these soils is 12% which is not statistically different from the pattern in the Rocky Flats soils. These data convincingly demonstrate that the leach procedure permits an accurate assessment of the plutonium content in many, if

Table 2

COMPLETE DISSOLUTION VERSUS LEACHING
Rocky Flats Soils

Map Site	Sample	Lab	Method	Pu-239 dpm/g ±% σ	% Deviation Between Methods
7	14	HASL	leach	3.08±4	3.2
		"	fusion	3.18±4	
6	6	HASL	leach	18.7±4	
		"	"	16.3±5	8.9
		"	fusion	16.0±5	
4	4	HASL	leach	0.060±4	28
		"	fusion	0.080±4	
				mean:	13.

Other Soils

Map Site	Sample	Lab	Method	Pu-239 dpm/g ±% σ	% Deviation Between Methods
N.Y.C. 6/67	0-20 cm	IPA	leach	0.0167±4	
		"	"	0.0165±5	0
		HASL	fusion	0.0166±4	
N.Y.C. 12/69	0-2.5 cm	TLW	leach	0.22±10	
		"	"	0.20±5	11
		"	HF dissolution	0.23±8	
		"	"	0.24±5	
N.Y.C. 12/69	0-5 cm	TLW	leach	0.091±6	
		"	"	0.096±5	3.3
		"	HF dissolution	0.090±6	
		"	"	0.092±5	
N.Y.C. 12/69	5-20 cm	TLW	leach	0.0048±20	
		"	"	0.0041±16	
		"	HF dissolution	0.0035±42	7.1
		"	"	0.0049±13	
		HASL	fusion	0.0041±5	
"Black" Soil, 1958		TLW	leach	0.0074±35	37.
		HASL	fusion	0.0051±4	
				mean:	12.

not all, soils and certainly in the soils encountered in the Rocky Flats area.

The deviations between methods reported in Table 2 include the error terms attendant to aliquoting and analytical precision. If the sample is not homogenized, deviations between aliquots will be high. By minimizing the error due to analytical precision, the data in Table (2) suggests that the error due to aliquoting is ±13% or less.

RESULTS

The Pu-238 and Pu-239 analyses of all the Rocky Flats samples plus additional reference samples are given in Tables A and B of the Appendix. Complete information regarding the collection area, the depth and weight of sample or fraction thereof, the analytical laboratory, and Pu-238/Pu-239 ratios are also reported. The mCi/km^2 of Pu-239 and Pu-238 integrated from 0 - 20 cm at each map site was calculated from Table A and is summarized in Table 3 along with the integrated Pu-238/Pu-239 ratio. When duplicate analyses were reported, the average of the duplicates were used in calculating the map site value unless otherwise specified in Table A. The deposition contours derived from Table 3 are plotted in Figure 5.

Since a number of earlier reports gave their data only in units of dpm/g soil, these values are also shown in Table A. These units are not at all useful for our study, and are difficult to compare without specifying depth of sampling and sample pretreatment.

The results of the depth profile studies are summarized in Table 4 as percent of the integrated activity down to 20 cm found in each incremental depth sampled. Profiles of fallout plutonium in New York City and Waynesville, Ohio are included for comparison.

*Footnote: From Table A it can be seen that at map site No. 8 the 13 - 20 cm increment contained more plutonium than the preceding increment. In evaluating this depth profile in Table 4 we summed the 9 - 13 cm and the 13 - 20 cm values and reported the total activity per unit area from 9 - 20 cm. Similarly at map site No. 6 we combined the 13 - 15 cm and 15 - 18 cm values for presentation in Table 4.

Table 3

TOTAL PLUTONIUM DEPOSITED AT SOIL SAMPLING SITES
IN THE ROCKY FLATS AREA

Map Site	mCi per mi^2 Pu-239	mCi per mi^2 Pu-238	Pu^{238}/Pu^{239}
1	2.4	0.07	0.029
2	3.1	0.06	0.019
3	4.2	0.12	0 028
4	11.	0.23	0.021
5	15	0 36	0 024
6	1950.	37.7	0.019
7	480.	8 9	0.018
8	630.	12.5	0.020
9	2.6	0.07	0.027
10	2.2	0.63	0 29
11	5.4	0.25	0.046
12	47.	1.2	0.026
13	50.	0 94	0.019
14	17.	0.39	0.023
15	18.	0.32	0.018
16	19.	0.41	0.022
17	14.	0.23	0.016
18	2.0	0.06	0.030
19	8.0	0.20	0.025
20	2.6	0.13	0.050
21	2.7	0.06	0.022
22	2.2	0.13	0.059
23	3.0	0.07	0.023
24	2.6	0.06	0.023
25	2.4	0.07	0.029
26	2.3	0.16	0.070
27	2.0	0.05	0.025
28	2.5	0.20	0.080
29	2.0	0.15	0.075
30	2.8	0.15	0.054
31	2.1	0.06	0.028
32	2.7	0.22	0.081
33	1.8	0.06	0.033

Figure 5:

PLUTONIUM-239 CONTOURS AROUND
ROCKY FLATS

Contour values in mCi per km^2
(light figures)
Numbered sampling sites
(bolder figures)
AEC property boundary -·-·-·-·
Dow Rocky Flats Plant --------

Table 4

DEPTH DISTRIBUTION OF PLUTONIUM
Rocky Flats Soils

Map Site	depth (cm)	% of total Pu^{239}
6	0-5	68
	5-8	24
	8-10	6
	10-13	1
	13-18	<1
8	0-6	66
	6-9	21
	9-20	13
8	0-5	67
	5-20	33
12	0-5	91
	5-20	9
14	0-5	61
	5-20	39
26	0-5	39
	5-20	61
27	0-5	45
	5-20	55
29	0-10	92
	10-20	8

Other Soils

Site	Sampling date		
New York City	Dec. 1969	0-2½	66
		0-5	81
		5-20	19
Waynesville, Ohio	Apr. 1970	0-6	47
		6-20	53

QUALITY CONTROL

HASL carries on an elaborate and extensive quality control program, both internally and with its contractors, to assure the validity of reported analyses. Since this particular study differs from our fallout programs, the quality control procedures and results are reported here in detail.

To monitor the quality of the analytical results, blind control samples were admixed with the normal schedule of routine samples. These controls included blank or low level Pu-239 samples, duplicate aliquots of prepared samples, and several reference samples.

Blank and Low level Samples

Two low level samples were used. One was a pre-bomb soil, that is a soil collected prior to 1945 which is the birth year of nuclear weaponry. A second was a soil sample retrieved from the town of Woodcliff Lake, New Jersey in March 1970 at a depth below 90 cm which was processed along with the other samples. The results of the analyses of these two samples are given in Table 5.

In general, the results in Table 5 show no detectable plutonium activity in the pre-bomb sample and a surprising value of about 0.005 dpm per gram Pu-239 for the Woodcliff Lake sample. Either a

Table 5

PLUTONIUM IN BLANK AND LOW LEVEL SAMPLES

Sample	Lab	dpm per gram Pu-239	dpm per gram Pu-238
Pre-bomb (collected before 1945)	HASL	0.0003 ± 100%	0.0002 ± 100%
"	IPA	0.0001 ± 100%	0.0001 ± 100%
"	IPA	0.0001 ± 100%	0.0001 ± 100%
"	TLW*	0.0196 ± 7%	0.0054 ± 14%
"	TLW	0.0001 ± 100%	0.0001 ± 100%
Woodcliff Lake, N.J.	IPA	0.0046 ± 7%	0.0001 ± 100%
(Collected below 90 cm	IPA	0.0043 ± 6%	0.0001 ± 100%
in March 1970)	TLW	0.0071 ± 9%	0.0009 ± 53%
	TLW*	0.0468 ± 5%	0.0001 ± 100%
	TLW	0.0055 ± 25%	0.0002 ± 100%

*Suspect value

small amount of Pu-239 penetrated to that depth or more reasonably the sample was slightly contaminated during collection or subsequent pre-analyses treatment. However, in one of the replicate analyses of each of these samples, Trapelo appears to have introduced from 2 to 4 dpm of Pu-239 contamination (100 gram aliquots were analyzed). This can represent a major portion of the total activity found in samples remote from the Rocky Flats plant; although the data in Table 5 also suggest that this contamination at Trapelo is not consistent. Examination of Table 5 and Tables A and B in the Appendix indicates that, on balance, contamination by laboratory handling and analyses was not measurable or was relatively unimportant to the outcome of this study.

Analytical Precision

The percent deviation between aliquots of a prepared sample, which represents the difference between aliquots divided by the mean, was calculated for each set of duplicates in Tables A and B. The data were subdivided into the internal replication at IPA, TLW, and HASL and also into the replication between these laboratories. Then the mean percent deviation between aliquots for each subgroup was computed and is reported in Table 6. It should be noted that the results of several samples reported by TLW were suspect, and duplicate aliquots of these samples were subsequently submitted to IPA for verification. These sets of duplicates were included in the compilation of Table 6 and reflect a slight bias against the random selection of duplicate sets.

Table 6 indicates that the precision of Pu-239 analysis both internally and between IPA and HASL ranged up to ±28% and averaged below ±20%. The internal precision at TLW and the precision between TLW and the other laboratories was 3 times higher. These uncertainties reflect not only the precision of analysis but also the representivity of aliquoting the prepared sample. For example, Table 2 suggests that aliquoting Rocky Flats soils gives an average uncertainty of less than 13%. Each of the results of the Rocky Flats soil analyses were examined in the light of expected results based upon location and comparison to neighboring samples. Reanalyses were performed when deemed necessary, and data were accepted depending upon their reasonableness and the laboratory conducting the analysis. Therefore, the overall analytical precision was about ±20%.

The level of Pu-238 in many of the samples was so low as to incur large counting errors. If either aliquot had a counting error

Table 6

MEAN PERCENT DEVIATION BETWEEN ALIQUOTS

Laboratory	Pu-239 No. of Sets	Pu-239 % Deviation
IPA	5	14
TLW	8	73
HASL	1	14
IPA/HASL	3	28
TLW/HASL	1	56
TLW/ IPA	9	58

greater than ±30%, that duplicate set was omitted from further consideration. With that limitation, the mean percent deviation between aliquots for Pu-238 analyses was very similar to the behavior of Pu-239.

Analytical Accuracy

The accuracy of the Pu-239 analyses is difficult to establish because of the lack of a standard soil sample. However, several reference samples were analyzed in which the plutonium levels can be approximated from their Sr-90 content. Table 7 identifies these samples giving the Sr-90 measurements, the expected Pu-239 values based upon a Pu-239/Sr-90 ratio of 0.02 [6], and the reported Pu-239 measurements. The "red" and "black" soils which were collected in 1958 at Raleigh, North Carolina and McHenry County, Illinois, respectively, were prepared as reference soil samples and were analyzed many times for Sr-90 content. The initial Pu-239 analyses of the "red" and "black" soils by TLW reflected the laboratory contamination discussed earlier. The re-analyses gave values which are about twice the expected values but in agreement with HASL's estimate of the "black" soil using the fusion treatment. The measured Pu-239 in the Derby, Colorado (1965) and New York City (1967)

Table 7

ANALYSES OF REFERENCE SAMPLES

Sample	Collection date	Sr-90	Expected[1] Pu-239	Measured Pu-239 (dpm per gram) TLW	IPA	HASL
		dpm/g ± Stnd Dev. %				
"Red Soil"	1958	0.14±6	0.0028	0.0567±8[3] 0.0061±14		
"Black Soil"	1958	0.18±4	0.0036	0.0399±9[3] 0.0074±35		0.0051±4[2]
Derby, Colorado	1965	0.43±7	0.0086	0.0110±7	0.0112±4	
New York City	1967	0.66±3	0.0132		0.0167±4 0.0165±5	

(1) Assuming Pu-239/Sr-90 = 0.02
(2) Analysis performed by fusion of sample
(3) Suspect value

samples are also higher than the expected values but by less than a factor of two. While these results cannot quantify the accuracy of analysis, they do demonstrate the acceptability of the method.

The laboratories performing these analyses were in good calibration with standard solutions of Pu-238 and Pu-239 made available by HASL. From these quality control studies it appears that the precision of replicate aliquoting and analysis is the determining factor in the uncertainty of the analytical results, and that the overall reliability of the analyses is within ±25%.

Representativity of Soil Sampling

To demonstrate that the sample collected was representative of the soil contamination at the sampling site, duplicate samples were collected by each of the two methods of sampling, and the results of these analyses are shown in Table 8. One sample of each duplicate sample was analyzed in replicate while the second duplicate was analyzed only once.

The template sampling method at map site 6 reflected a ±31% deviation between duplicate samples for Pu-239 while the coring method at map site 7 showed only a ±11% deviation. The Pu-238 results were similar because the Pu-238/Pu-239 ratio for all samples

Table 8

DUPLICATE SOIL SAMPLING IN THE ROCKY FLATS AREA

Map Site	depth (cm)	Sampling method	No. of analyses	mCi per km^2 Pu-239	mCi per km^2 Pu-238	Pu^{238}/Pu^{239}
6	0-20	Template	4	2050	40.3	0.020
			1	1500	27.4	0.018
7	0-20	Cores	3	490	9.4	0.019
			1	437	7.5	0.017

was remarkably constant. This precision is not unlike the precision of replicate analyses of identical samples. We conclude that the representivity of soil sampling is good and that the uncertainty of the final data is due largely to the imprecision of aliquoting and analysis.

D I S C U S S I O N

Distribution with depth

Significant quantities of Pu-239 are below the top 5 cm of soil and vegetation. Table 4 shows that between 9 and 61 percent of the Pu-239 from the surface to 20 cm in depth is below 5 cm, depending upon the particular type of soil. At map sites nos. 6 and 8 we sampled three or more increments to see how the plutonium was distributed with depth. Although about 90% was in the top 9 cm we were able to measure Pu-239 as far down as 13 cm. We would conclude, however, that at these sites less than 1% is below this depth.

At five other sites we separated the 0 - 5 cm and 5 - 20 cm fractions and found widely varying percentages of the total Pu-239 measured, below 5 cm. Undoubtedly the base exchange capacity and perhaps other characteristics of the soil are important factors in determining the extent to which deposited plutonium will penetrate below the surface. Sites Nos. 26 and 27 were in city parks where liming and fertilization may enhance the downward movement. Lacking better definition of the plutonium distribution with depth at these other sites we assumed that in sampling to 20 cm we removed essentially all of the plutonium that had fallen out.

Inventory

The areas within the various contours of Figure 5 were determined by planimetry, and the total Rocky Flats Pu-239 burden in the soil bounded by the 3 mCi/km^2 contour is estimated to be 4.5 Ci. One objective of this study was to determine the quantity of Pu-239 released to property not controlled by the AEC. The sampling plan reflected this objective by collecting few samples on the plant site. Consequently, our estimates of the Pu-239 within the plant area is poor and may be low. The Rocky Flats Pu-239 on public and private lands not controlled by the AEC and within the 3 mCi/km^2 contour is estimated to be 2.6 Ci.

There is no question that Rocky Flats plutonium extended beyond the 3 mCi/km^2 contour, but the extent of this low level contamination is difficult to define. Table B shows that the fallout Pu-239 in 1965 at Derby, Colorado which is only 17 miles east of Rocky Flats was 1.5 mCi/km^2. Additional nuclear weapons fallout from 1965 to 1970 would add little Pu-239 to this value. The observed values of

2.0 and 1.8 mCi/km^2 at map site Nos. 29 and 33 which were about 40 miles to the east and north of Rocky Flats, respectively, began to approach the expected fallout level of 1.5 mCi/km^2. The intervening sites were somewhat higher and averaged about 2.5 mCi/km^2. Taking the extreme and unlikely view that the entire area was contaminated with the difference (1 mCi/km^2) indicates that, as an upper limit, an additional 3.2 Ci of Pu-239 could have been released from the plant to the remote areas beyond the 3 mCi/km^2 contour. The total released to non AEC controlled property would then increase to 5.8 Ci.

This calculation of the remote contamination relies heavily upon the 1.5 mCi/km^2 estimate of fallout plutonium in the area. The one measurement at Derby in 1965 is not sufficient for a sound argument. Sr-90 analyses of the remote site samples and other studies are in progress which might resolve the problem. However, a thorough solution may require additional sampling in the Colorado area.

The Pu-238 measurements serve little useful purpose except to document the Pu-238/Pu-239 ratio of the Rocky Flats debris. The average ratio of this debris is about 0.020. At the remote sites where there is a greater proportion of fallout plutonium, the ratio is higher reflecting the debris from the 1964 re-entry burn-up of the SNAP-9A generator.

Source of Contamination

The distribution of Rocky Flats plutonium within the 3 mCi/km^2 contour suggests that the leaking barrels were the source of the contamination. The average wind behavior over the 17 years of record (Figure 1) should distribute a chronic low-level stack effluent in a directorial pattern similar to the contamination contours shown in Figure 5. However, the downwind extent of the stack effluent would depend largely on the particle size of the plutonium. Even if only gravitational settling is considered for the PuO_2 emitted from the 250 foot stack at the Dow plant, and if an average westerly wind of 12 miles per hour is assured (Figure 1), then the plutonium within the 3 mCi/km^2 contour must have had a particle size in excess of 10μ diameter. The particle size distribution of the plutonium influent to the stack is not available, although measurements are in progress to determine it (4). Moreover, stack monitoring from the start of plutonium operations in 1953 through 1968 indicate an integrated release of only 38 mCi (7) of plutonium - far below what we found.

The May 11, 1969 fire can be exonerated as the source because the winds on that day (Figure 2) were almost directly opposite to the contamination pattern leaving the plant site (Figure 5). The detection of plutonium at 13 cm below the soil surface would further tend to rule out a significant contribution from this event. The September 11, 1957 fire can not be eliminated on the basis of the winds at the time of that fire (Figure 3). However the particle size of the dioxide from burning plutonium is estimated to be submicron (2). The sharp gradient and relatively short downwind extent of the contours in Figure 5 are not compatible with deposition of sub-micron particles.

There are several observations about the plutonium distribution which link the contamination to the leaking barrels. The plutonium laden oil from the corroded barrels contaminated the soil in and around the storage area. As the oil seeped underground or evaporated, strong gusty winds which occur predominantly from the west, resuspended the contaminated soil carrying it eastward. Because the efficiency for resuspending particulates decreases with lower wind velocities, the relatively light easterly winds would not distribute much contamination to the west of the plant site.

The barrel storage area was in a region on Rocky Flats where the ground was mostly course rock and sand particles. If these particles are resuspended they would fall back to the ground rather quickly. The rapid decrease of the high level contours east of the plant site supports this view. Similarly the contamination did not concentrate in the creek ravines as do the fine particles of soil and alluvium.

It is interesting and probably meaningful that the surveys of the barrel storage area on the plant site suggested a southeasterly migration of the contamination. The contours off the plant site in Figure 5 also show a strong southerly component.

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Finally we acknowledge the contribution of Norton Chu who developed the analytical and assay procedure for plutonium in soil.

Table A

ANALYTICAL RESULTS : Rocky Flats Soils

Map Site	Sample	Sampling date	depth (cm)	area (cm²)	air dried wt. - kg total	rocks	soil	anal. lab	dpm per gram dried soil(5) Pu-239	Pu-238	mCi per km² Pu-239	Pu-238	Pu-238 / Pu-239
1	1	2/11/70	0-20	966	38 29	17 60	20 69	IPA	0 0240 ± 3%	0 0008 ± 25%	2 3	0 08	0 033 ± 25%
2	2	2/11/70	0-20	1050	44 58	20 20	24 38	IPA	0 0295 ± 3%	0 0006 ± 25%	3 1	0 06	0 020 ± 25%
3	3	2/11/70	0-20	1003	35 94	14.38	21.56	IPA	0 0433 ± 3%	0 0012 ± 20%	4 2	0 12	0 028 ± 20%
4	4	2/10/70	0-20	622	18 44		18.44	HASL	0 0600 ± 4%	0 0019 ± 5%	8 0	0 25	0 031 ± 6%
								HASL (1)	0 0800 ± 4%	0 0013 ± 8%	10 7	0 18	0 016 ± 9%
								IPA	0 102 ± 2%	0 0020 ± 15%	13 6	0 27	0 020 ± 15%
5	5	2/10/70	0-15	622	16.46	0 85	15.61	IPA	0 134 ± 2%	0 0032 ± 9%	15 1	0 36	0 024 ± 10%
6	6	2/10/70	0-20	929	33 83	8.73	25.10	HASL	18 7 ± 4%	0 336 ± 4%	2270	40 9	0 018 ± 5%
								HASL(1)	16 0 ± 5%	0 352 ± 4%	1950	42 8	0 022 ± 6%
								HASL	16 3 ± 5%	0 318 ± 6%	1982	38 7	0 020 ± 7%
								TLW(2)	22 4 ± 3%	0 394 ± 6%	2725	47 9	0 018 ± 7%
								TLW(2)	39 8 ± 3%	0 866 ± 4%	4842	105	0 022 ± 5%
								IPA	16 6 ± 2%	0 318 ± 4%	2014	38 7	0 019 ± 5%
6	7	2/15/70	0-20	1003	27.29	5.89	21.40	TLW	15.6 ± 2%	0 285 ± 4%	1498	27 4	0 018 ± 5%
6	8	2/14/70	0-5	929			1.72	IPA	171 ± 3%	3 35 ± 3%	1425	27 9	0 020 ± 4%
								IIA	151. ± 3%	2 64 ± 3%	1260	22 0	0 018 ± 4%
6	9	2/14/70	5-8				1.35	IPA	35 8 ± 1%	0 659 ± 4%	402 (3)	8.7(3)	0 018 ± 5%
6	10	2/14/70	8-10				1.77	IPA	8 50 ± 6%	0 180 ± 6%	119 (3)	2 5(3)	0 021 ± 8%
6	11	2/14/70	10-13				1.10	IPA	1.20 ± 4%	0 0216 ± 4%	16 (3)	0 3(3)	0 018 ± 6%
6	12	2/14/70	13-15				0.78	IPA	0 287 ± 2%	0 0056 ± 8%	4 (3)	0 08(3)	0 020 ± 9%
6	13	2/14/70	15-18				0 63	IPA	0.290 ± 2%	0.0053 ± 10%	8 (3)	0 1(3)	0 018 ± 11%
7	14	2/10/70	0-20	622	19.81		19 81	HASL	3.08 ± 4%	0 059 ± 3%	442	8.47	0 019 ± 4%
								HASL(1)	3.10 ± 4%	0.066 ± 4%	456.	9 46	0 021 ± 5%
								IPA	4.00 ± 1%	0.072 ± 3%	474.	10 3	0 018 ± 4%
7	15	2/18/70	0-20	622	19.19		19.19	IPA	3.15 ± 1%	0 054 ± 5%	437	7.50	0.017 ± 6%
8	16	2/10/70	0-5	622	2.31		2.31	TLW	25.4 ± 4%	0.506 ± 5%	424	8 42	0 020 ± 6%
8	17	2/10/70	5-20	622	17 48		17 48	TLW	1 64 ± 3%	0 0324 ± 6%	207	4 09	0 020 ± 6%
8	18	2/15/70	0-6	81	0 39		0 39	TLW	11 6 ± 3%	0 350 ± 3%	252	7 61	0 030 ± 4%
								TLW	13.0 ± 3%	0.399 ± 4%	282.	8 64	0 031 ± 5%
8	19	2/15/70	6-9	81	0.22		0.22	TLW	6.72 ± 3%	0 131 ± 4%	82 3	1.60	0 020 ± 4%
8	20	2/15/70	9-13	46	0.22		0 22	TLW	0.355 ± 3%	0 0080 ± 10%	7 7	0 17	0 022 ± 11%
8	21	2/15/70	13-20	62	0.87		0.87	TLW	0.745 ± 3%	0.0157 ± 8%	46 8	0.99	0 021 ± 8%

ANALYTICAL RESULTS: Rocky Flats Soils cont'd

Table A - cont'd

sep site	sample	Sampling date	depth (cm)	area (cm²)	air dried wt.-kg total	rocks	soil	anal lab	dpm per gram dried soil Pu-239	Pu-238	mCi per km² Pu-239	Pu-238	Pu-238/Pu-239
9	22	2/11/70	0-20	929	20.41	2.58	17.83	IPA	0.0320±3%	0.0007±15%	2.8	0.08	0.027±15%
								IPA	0.0266±1%	0.0008±30%	2.3	0.07	0.030±30%
10	23	2/12/70	0-20	966	22.99	1.65	21.34	TLW	0.0220±5%	0.0063±10%	2.2	0.62(2)	0.29± 1%
11	24	2/10/70	0-20	622	17.60		17.60	IPA	0.0420±1%	0.0020±100%	5.4	0.25	0.048±100%
12	25	2/9/70	0-5	622	2.98		2.98	TLW	2.14±3%	0.0409±5%	46.2	0.88	0.019±6%
								IPA	1.83±3%	0.0401±6%	39.5	0.86	0.022±7%
12	26	2/9/70	5-20	622	14.28		14.28	TLW	0.0443±4%	0.0053±11%	4.6	0.54	0.12±12%
								IPA	0.0361±5%	0.0010±40%	3.7	0.10	0.028±40%
12	27(4)	2/9/70	0-10										
13	28	2/11/70	0-20	1003	21.88	1.76	20.12	IPA	0.559±3%	0.0101±11%	50.5	0.94	0.019±12%
14	29	2/11/70	0-5	622	2.82		2.82	TLW	0.517±5%	0.0113±16%	10.5	0.23	0.022±16%
								IPA	0.510±3%	0.0105±10%	10.4	0.21	0.020±11%
14	30	2/11/70	5-20	622	13.94		13.94	TLW	0.109±4%	0.0021±28%	10.1	0.24	0.024±28%
								IPA	0.0332±5%	0.0010±30%	3.4	0.10	0.030±30%
15	31	2/12/70	0-20	929	26.41		26.41	IPA	0.138±4%	0.0025±33%	17.7	0.32	0.018±30%
16	32	2/13/70	0-20	622	14.52		14.52	TLW(2)	0.483±3%	0.0113±9%	50.7	1.19	0.023±10%
								IPA	0.182±3%	0.0039±15%	19.2	0.41	0.021±15%
17	33	2/12/70	0-20	1003	36.56	12.20	24.36	IPA	0.126±2%	0.0021±17%	47.0	0.23	0.017±17%
18	34	2/13/70	0-20	622	16.08		16.08	TLW	0.0176±7%	0.0005±90%	2.0	0.06	0.028±100%
19	35	2/13/70	0-20	622	15.90		15.90	IPA	0.0698±5%	0.0017±30%	8.0	0.20	0.024±30%
20	36	2/12/70	0-20	1003	22.47		22.47	IPA	0.0262±4%	0.0013±30%	2.6	0.13	0.050±30%
21	37	2/13/70	0-20	622	16.68		16.68	IPA	0.0180±4%	0.0004±100%	2.3	0.05	0.021±100%
								IPA	0.0253±3%	0.0007±20%	3.0	0.08	0.028±20%
22	38	2/13/70	0-20	622	17.17		17.17	TLW	0.0174±7%	0.0010±10%	2.2	0.13	0.057±48%
23	39	2/14/70	0-20	622	16.41		16.41	IPA	0.0254±6%	0.0006±100%	3.0	0.07	0.024±100%
24	40	2/14/70	0-20	622	16.52		16.52	IPA	0.0220±7%	0.0005±100%	2.6	0.06	0.023±100%
25	41	2/14/70	0-20	622	16.09		16.09	IPA	0.0205±6%	0.0006±100%	2.4	0.07	0.029±100%
26	42	2/12/70	0-5	622	3.55		3.55	TLW	0.0343±4%	0.0022±25%	0.88	0.06	0.064±25%
26	43	2/12/70	5-20	622	17.61		17.61	TLW	0.0136±7%	0.0009±50%	1.7	0.12	0.066±50%
								IPA	0.0085±6%	0.0007±40%	1.1	0.09	0.082±40%

Table A - cont'd

ANALYTICAL RESULTS: Rocky Flats Soils cont'd

Map Site	Sample	Sampling date	depth (cm)	area (cm^2)	air dried wt. - kg total	air dried wt. - kg rocks	air dried wt. - kg soil	anal. lab	dpm per gram dried soil Pu-239	dpm per gram dried soil Pu-238	mCi per km^2 Pu-239	mCi per km^2 Pu-238	Pu-238 / Pu-239
27	44	2/12/70	0-5	522	2.72		2.72	IPA	0 0453 ± 3%	0.0010 ± 20%	0.80	0 02	0 022 ± 20%
27	45	2/12/70	5-20	622	13 94		13 94	IPA	0 0107 ± 5%	0.0003 ± 80%	1 1	0 03	0 028 ± 40%
28	46	2/13/70	0-20	622	20 69		20.69	TLW	0 0166 ± 6%	0.0013 ± 35%	2 5	0 20	0 078 ± 35%
29	47	2/12/70	0-10	622	6 04		6.04	IPA	0 0422 ± 3%	0 0018 ± 20%	1 8	0 08	0 042 ± 20%
29	48	2/12/70	10-20	622	13 90		13.90	IPA	0.0016 ± 14%	0.0007 ± 30%	0 16	0.07	0 44 ± 30%
30	49	2/13/70	0-20	678	22 08		22.08	TLW	0 0190 ± 7%	0 0010 ± 57%	2 8	0 15	0 053 ± 57%
31	50	2/13/70	0-20	622	17.44		17.44	IPA	0 0170 ± 4%	0 0005 ± 30%	2.1	0.06	0 029 ± 30%
32	51	2/13/70	0-20	622	21.34		21.34	TLW	0 0192 ± 6%	0 0022 ± 26%	3 0	0 34	0 11 ± 26%
								TLW(2)	0 734 ± 2%	0.0134 ± 7%	113.	1 1	0 018 ± 7%
								IPA	0 0153 ± 5%	0.0006 ± 100%	2.4	0 09	0.039 ± 100%
33	52	2/13/70	0-20	622	16.94		16.94	IPA	0.0144 ± 4%	0.0005 ± 30%	1.8	0 06	0 035 ± 30%

(1) sodium carbonate fusion

(2) plutonium results not used in averaging

(3) calculated from estimates of soil density

(4) sample not analyzed

(5) 100 gram aliquot of air dried soil leached by HASL method

Table B

ANALYTICAL RESULTS: Reference Soils

Reference Soil	Sampling date	depth (cm)	area (cm²)	Total air-dried weight (kg)	anal lab	dpm per gram dried soil(3) Pu-239	dpm per gram dried soil(3) Pu-238	mCi per km² Pu-239	mCi per km² Pu-238	Pu238/Pu239
Derby, Colo.	Sept 1965	0 - 20	622	18.30	IPA	0.0112 ± 4%	0.0003 ± 30%	1.5	0.04	0.027 ± 30%
					TLW	0.0110 ± 7%	0.0041 ± 15%	1.4	0.54(2)	0.37 ± 17%
New York, N.Y.	June 1967	0 - 20	622	17.55	IPA	0.0167 ± 4%	0.0004 ± 30%	2.1	0.05	0.024 ± 30%
					IPA	0.0165 ± 5%	0.0003 ± 30%	2.1	0.04	0.018 ± 30%
					HASL(1)	0.0166 ± 4%		2.1		
New York, N.Y.	Dec 1969	0 - 2.5	186	0.24	TLW(4)	0.22 ± 10%	0.012 ± 63%	1.3	0.07	0.053 ± 63%
					TLW(4)	0.20 ± 5%	0.012 ± 25%	1.2	0.07	0.057 ± 25%
					TLW(5)	0.23 ± 6%	0.011 ± 45%	1.4	0.06	0.047 ± 45%
					TLW(5)	0.24 ± 5%	0.013 ± 28%	1.4	0.08	0.053 ± 28%
New York, N.Y.	Dec 1969	0 - 5	622	2.41	TLW(4)	0.091 ± 6%	0.0006 ± 100%	1.6	0.02	0.005 ± 100%
					TLW(4)	0.096 ± 5%	0.0078 ± 25%	1.7	0.14	0.086 ± 25%
					TLW(5)	0.090 ± 6%	0.0040 ± 52%	1.6	0.07	0.044 ± 52%
					TLW(5)	0.092 ± 5%	0.0028 ± 68%	1.6	0.05	0.031 ± 69%
					HASL(1)	0.093 ± 4%		1.6		
New York, N.Y.	Dec 1969	5 - 20	622	11.86	TLW(4)	0.0048 ± 20%	0.0087 ± 22%	0.41	0.74	1.8 ± 22%
					TLW(4)	0.0011 ± 16%	0.0021 ± 77%	0.36	0.18	0.50 ± 77%
					TLW(5)	0.0035 ± 42%	0.0009 ± 100%	0.31	0.08	0.26 ± 100%
					TLW(5)	0.0049 ± 13%	0.0004 ± 100%	0.43	0.03	0.09 ± 100%
					HASL(1)	0.0011 ± 5%	0.0003 ± 67%	0.35	0.03	0.07 ± 67%
"Black" Soil	Nov 1958	0 - 5			TLW(2)	0.0399 ± 9%	0.0012 ± 30%			0.11 ± 30%
					TLW	0.0074 ± 35%	0.0036 ± 62%			0.49 ± 62%
					HASL(1)	0.0051 ± 4%	0.0002 ± 100%			0.04 ± 100%
"Red" Soil	Nov 1958	0 - 5			TLW(2)	0.0567 ± 8%	0.0016 ± 95%			0.03 ± 95%
					TLW	0.0061 ± 14%	0.0005 ± 123%			0.08 ± 123%
Waynesville, O.	Apr 1970	0 - 6	622	3.60	TLW	0.0440 ± 4%	0.0064 ± 11%	1.1	0.17	0.14 ± 11%
					IPA	0.0420 ± 3%	0.0020 ± 30%	1.1	0.05	0.05 ± 30%
Waynesville, O.	Apr 1970	6 - 20	622	14.35	TLW	0.0132 ± 7%	0.0011 ± 44%	1.4	0.11	0.08 ± 44%
					IPA	0.0109 ± 10%	0.0005 ± 100%	1.1	0.05	0.05 ± 100%

(1) Sodium carbonate fusion
(2) value suspect
(3) 100 gram aliquot of air dried soil leached by HASL method
(4) " " " " " " " " TLW "
(5) " " " " " " " completely dissolved in acid and HF.